SEASONALITY AND SOURCES OF LIGHT-ABSORBING AEROSOLS AT SUMMIT, GREENLAND

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SEASONALITY AND SOURCES OF LIGHT-ABSORBING AEROSOLS AT SUMMIT, GREENLAND

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LIST OF SYMBOLS AND ABBREVIATIONS

σ_{ap}	Particle light absorption coefficient
σ_{sp}	Particle light scattering coefficient
ω_o	Single scattering albedo
AAE	Absorption Ångström exponent
BC	Black carbon
BrC	Brown carbon
EPA	Environmental protection agency
GIS	Greenland ice sheet
TAWO	Temporary atmospheric watch observatory
GEO	Greenland environmental observatory
NSF	National science foundation
CLAP	Continuous light absorption photometer
PSAP	Particle soot absorption photometer
NOAA	National oceanic and atmospheric administration
NILU	Norwegian institute for air research
MSA	Methanesulfonic acid

SUMMARY

The Greenland ice sheet (GIS) is a key component of the warming Arctic climate, having the potential to dramatically influence sea level through melting. Light-absorbing aerosols are thought to be significant contributors to warming in the Arctic, because of their effect on the radiation balance through both aerosol absorption in the atmosphere as well as absorption in surface snow after particulate deposition. At this time it is not possible to estimate the impact of aerosol absorption on the radiation balance over Greenland due to the lack of in-situ measurements. Here, we present time series and estimates of key aerosol optical properties in order to better understand the seasonality and sources of aerosols over central Greenland, and compare their values with other Arctic sites. In-situ measurements made at Summit, Greenland from May 8, 2011 to December 31, 2014 include aerosol light absorption coefficient (σ_{ap}) and light scattering coefficient (σ_{sp}); calculated parameters include absorption Ångström exponent (AAE), and single scattering albedo (ω_o). The light absorption and scattering coefficients were found to be low in the winter and highest in the spring and summer. Spring-summer means of σ_{ap} and σ_{sp} were 0.15 ± 0.15 Mm^{-1} and 2.35 ± 2.80 Mm^{-1}, respectively. Mean AAE was 0.97 ± 0.29 in the spring and summer, indicating that black carbon (BC), and not dust and/or organic brown carbon (BrC), is the main aerosol light absorber. Mean ω_o was 0.93 ± 0.03, which is similar to values measured at Barrow, Alaska, USA (0.94 ± 0.05) and Ny-Ålesund, Svalbard, Norway (0.95 ± 0.06). Summit exhibits ω_o as low as Barrow and Ny-Ålesund although it is an isolated high-altitude site indicating the importance of aerosol light absorption over the most remote Arctic locations.

CHAPTER 1

INTRODUCTION AND BACKGROUND

1.1 A Brief Overview of Climate Change

The Earth's climate is changing. Over the past century, the global mean surface temperature has risen by approximately 0.74°C, and over the last 50 years, temperatures have risen at a rate of about 0.13°C per decade (Trenberth *et al.*, 2007). The warming of the Earth has been attributed mainly to an increase in carbon dioxide (CO_2) and other greenhouse gases which are largely produced from fossil fuel combustion. In addition to rising temperatures, changes in the weather and climate have also been observed. The United States Environmental Protection Agency (EPA) reports that the total global annual precipitation over land areas has increased by 0.5% since 1901. It also reports increased heat waves in the US in the past century with the 1930s experiencing the most severe heat waves. In addition, the last few decades in the US have seen an increase in unusually hot summer days and nights (USEPA, 2014). Global sea level has risen 19.5 cm from 1870 to 2004 at an approximate rate of 0.17 cm per year (Church and White, 2006). The Arctic has also been affected by the Earth's changing climate, experiencing decreased sea ice and snow cover (Mote, 2007).

1.2 Aerosols in the Arctic

The Arctic region is warming more quickly than other parts of the Earth. Studies show that in the past 50 years, the Arctic has warmed at twice the average global rate.

(Trenberth *et al.*, 2007; Walsh, 2014). Light-absorbing aerosols, especially black carbon (BC), have been estimated to be a significant contributor to this warming over the past 30 years. Shindell and Faluvegi (2009) compare model estimates of aerosol radiative forcing with historical emissions data and found that warming occurred when BC emissions increased and sulfate aerosols decreased. Light-absorbing aerosols contribute to atmospheric warming by shifting the energy balance to a net positive radiative forcing. This can be achieved through absorbing solar radiation directly and by decreasing the surface albedo after deposition. The primary light-absorbing aerosols in the Arctic are BC, organic brown carbon (BrC) and dust. The Arctic is particularly susceptible to regional radiative forcing by aerosols due to the prevalence of high-albedo snow and ice surfaces.

1.3 Aerosols over the Greenland Ice Sheet

A key component of the Arctic climate is the highly reflective Greenland ice sheet (GIS). The GIS is the largest glacial ice mass in the Arctic and the second largest on Earth, covering an area of approximately 1.71 million km^2 and a volume of 2.85 million km^3. The amount of freshwater stored in the ice sheet has the potential to increase sea level by more than 7 m if it were to melt completely, altering ocean circulation and global climate. Studies indicate that the entire surface of the ice sheet may melt on a seasonal basis within 10 years (Box *et al.*, 2012) and could potentially contribute to a 32 cm rise in sea level within 40 years (Rignot *et al.*, 2011). The GIS also exhibits an extremely high surface albedo due to the vast amount of highly reflective snow cover and sea ice. As a result, it plays a crucial role in the regional energy balance and substantially impacts

regional and global climate. Furthermore, much of the GIS is at high altitude, causing it to experience different transports of aerosols and meteorological patterns than other parts of the Arctic.

Evidence suggest that light-absorbing aerosols may be causing significant warming over the GIS. A study, using remote sensing data, proposes that deposited aerosols have significantly lowered the springtime surface albedo of the GIS since 2009, leading to a surface mass loss of 27 Gt yr^{-1} (Dumont *et al.*, 2014). Another study shows that widespread melting on the ice sheet is driven by the combination of decreased surface albedo and simultaneous periods of warm temperatures which likely caused the melting events observed in 1889 and 2012 (Keegan *et al.*, 2014). Evidence also suggests that low-level 'liquid clouds' may be enhancing warming over the GIS. Bennartz *et al.* (2013) show that during the 2012 melt event, the presence of these clouds caused near-surface temperatures to rise above the melting point. This finding further supports the significance of light-absorbing aerosols, because they can act as cloud condensation nuclei for cloud formation (Breider *et al.*, 2014).

1.4 Key Optical Properties

Key aerosol optical properties used to analyze the impact of light-absorbing aerosols on climate include the aerosol light absorption coefficient (σ_{ap}), the aerosol light scattering coefficient (σ_{sp}), the absorption Ångström exponent (AAE), and the single scattering albedo (ω_o). The light absorption coefficient is used to quantify how effectively particles absorb sunlight or solar radiation. It can also be used to indicate the presence and amount of light-absorbing aerosols. Treated independently, the absorption of solar

radiation by aerosols results in warming of the planet. The light scattering coefficient is used to quantify how effectively particles scattering sunlight or solar radiation. The scattering of solar radiation by aerosols results in cooling of the planet. The single scattering albedo measures the fraction of light extinction (sum of absorption and scattering) that is due to scattering. It is one of the most important properties of aerosols as it controls whether an aerosol layer will have a net cooling or warming effect. A ω_o of one indicates a completely scattering aerosol whereas a low value indicates a more absorption. It plays a key role in determining the impact aerosols have on the radiation balance. The absorption Ångström exponent measures the wavelength dependence of the absorption coefficient which is distinct for each aerosol. The AAE can be used to identify and distinguish between different types of aerosols.

1.5 Overview of Study

It is likely that aerosols are important elements to Arctic warming, however, their seasonality over central Greenland, particularly during the summer when solar insolation is at its peak, is not well understood. While some studies (e.g. Hagler *et al.*, 2007) do present estimates of the aerosol light absorption coefficient for the region, they are limited to only a single year or season. The literature currently lacks continuous, interannual measurements of the aerosol optical properties over central Greenland needed to characterize seasonality and inter-annual variability. This paper presents these measurements including aerosol light absorption and scattering coefficients, absorption Ångström exponent, and single scattering albedo at Summit, Greenland spanning from May 8, 2011 to December 31, 2014. These measurements help to identify periods of high

absorption which reveal seasonal and annual variability, to distinguish the primary source of light absorption from BC, dust, or brown carbon (BrC) when absorption is high using AAE, and to assess the importance of light-absorbing aerosols over central Greenland using ω_o.

CHAPTER 2

METHODOLOGY

2.1 Site Description

The data presented in this paper were collected at Summit, Greenland, located at 72°N, 38°W, with an elevation of approximately 3200 m above sea level. Sampling took place at Summit's Temporary Atmospheric Watch Observatory (TAWO) located within a designated clean air sector approximately one kilometer south from the main camp at the Greenland Environmental Observatory (GEOSummit). The site is located in the center of Greenland, a clean, isolated, high-altitude area that serves as a year-round sampling station funded by the National Science Foundation (NSF). Sources of contamination are still present, however, from sources such as the camp's diesel generator as well as from vehicle and flight traffic. Procedures are taken to limit these emissions during periods of north winds to prevent as much contamination as possible.

2.2 Instrumentation

The instruments used in this this study consisted of a TSI nephelometer and a Continuous Light Absorption Photometer (CLAP) developed by the National Oceanic and Atmospheric Administration (NOAA) (Ogren *et al.*, 2013). The CLAP is a multi-spot version of the Radiance Research Particle Soot Absorption Photometer (PSAP). The nephelometer measured aerosol light scattering coefficient at three different wavelengths (450, 550, and 700 nm) with an uncertainty of ±7% (Delene and Ogren, 2002). Light

absorption coefficient was measured at three different wavelengths (467, 528, and 652 nm) with the CLAP which has an uncertainty of ±2% (Ogren *et al.*, 2013). Standard corrections for instrument non-idealities (e.g. nephelometer truncation, etc.) were applied to these data as described in Delene and Ogren (2002). The CLAP data are treated as PSAP data in terms of corrections. Values of σ_{ap} and σ_{sp} are reported at standard temperature (T = 0°C) and pressure and at low relative humidity (RH<40%). The CLAP-measured σ_{ap} were adjusted to the nephelometer wavelengths by interpolation using the absorption Ångström exponent. AAE can be calculated as in the following:

$$AAE = -\frac{log[\sigma_{ap,652}/\sigma_{ap,467}]}{log[652nm/467nm]}$$

(1)

where $\sigma_{ap,652}$ is the light absorption coefficient measured at one wavelength of interest (652 nm) and $\sigma_{ap,467}$ is the light absorption coefficient measured at another (467 nm). For our analysis, AAE was calculated from 450 nm to 700 nm, after the CLAP wavelength adjustments were made. Uncertainty in AAE is ±2.8% based on the propagation of measurement uncertainty of σ_{ap} (±2%) from the CLAP.

Single-scattering albedo (ω_o) was calculated at 550 nm using the measured σ_{sp} and σ_{ap} from the nephelometer and CLAP, respectively, and the following equation:

$$\omega_o = \frac{\sigma_{sp}}{\sigma_{sp}+\sigma_{ap}}$$

(2)

Uncertainty in ω_o is ±10.1% based on the propagation of measurement uncertainty of σ_{sp} (±7%) from the nephelometer and σ_{ap} (±2%) from the CLAP.

Single-scattering albedo at 550 nm was also calculated for Barrow, Alaska, USA and Ny-Ålesund, Svalbard, Norway using σ_{sp} and σ_{ap} measurements from the Norwegian Institute for Air Research (NILU) EBAS database. The Barrow dataset spans from January 1, 2009 to December 31, 2013, and the Ny-Ålesund dataset spans from January 1, 2005 to December 31, 2009.

2.3 Quality Control Procedures

The data at Summit was collected using the NOAA-developed software LiveCPD which runs on the on-site computers and can also be used to control the instruments. The data can then be reviewed, processed, analyzed, and archived using NOAA's AER_VM software where the data is presented graphically. The data used in this study was manually processed for quality control using the AER_VM software. A detailed procedure for the process is as follows:

1. The AER_VM software is accessed through a virtual machine player (e.g. VMware player).
2. At the home screen inside the software, the data from the user computer must be synchronized with data from the field computers.
3. After synchronization, the command terminal is opened. The command "data.status *stn*" is entered to see the date of the last data received and the date of the last data passed as clean data, where "*stn*" is the 3-character station code ("sum" for Summit).

4. The command “cpx2 *stn YYYY*w*WW N*w” is entered to load and view the data to be edited where *YYYY* is the 4-digit year, *WW* is the 2-digit week, and *N* is the number of weeks to be loaded.
5. In the cpx2 window where the data is shown, the “Edit” view is selected from the “Display” dropdown window.
6. Available here are data for scatter and backscatter from the nephelometer, absorption from the PSAP and CLAP, wind speed, wind direction, etc.
7. The message log is opened through the “CPX2” dropdown window and can be viewed to help explain data behavior
8. The “Mentor edits” window is opened through the “CPX2” dropdown window and is used to make an edit.
9. To make an edit, the target data is zoomed in to and highlighted and “Add” is selected to open the “Edit Directive”.
10. In the “Edit Directive” window, the target dataset or group of datasets are selected, the option to mark the data as “invalidate” or “contaminate” is selected, and information regarding the author and edit notes are imputed.
11. “Invalidate” is selected if the data shows abnormal variability, a sampling or measurement problem is determined, or the data is physically unrealistic. “Contaminate” is selected if local aerosol sources caused the data to be not representative of the regional or target aerosol.
12. “OK” and then “Save” is selected to save the edit.
13. Common edits include when there are data spikes (generally short-duration deviations from normal measurements often caused by local contamination or

measurement problems) and when the data is during periods of north winds or stagnant winds.

14. Once all edits are made and saved, the data can be closed, and the data can be passed as clean.
15. To pass the data as clean, the command “data.pass *stn YYYY*w*WW N*w” is entered into the command terminal.
16. Repeat these steps to edit additional weeks of data.

CHAPTER 3

RESULTS AND DISCUSSION

3.1 Absorption and Scattering

3.1.1 Absorption

Monthly statistical distributions of the aerosol properties based on daily-averaged data from the sample period are analyzed. Statistical distributions are represented as box-whisker plots (Fig. 1,2,4,6) where the whiskers represent the 5 and 95 percentiles, the bottom and top of the box represent the 25 and 75 percentiles, and the marker inside the boxes represents the median values. Fig. 1 shows the monthly values of absorption coefficient at Summit over the sampling period. Absorption is highest during spring (April, May, June) and summer (July, August), when the sun is out and light absorption is most significant. The CLAP measured an average σ_{ap} (at 550 nm) of 0.15 ± 0.15 Mm^{-1} over these months from 2011 to 2014. In a previous study, Hagler *et al.* (2007) found σ_{ap} at Summit during the summer of 2006 to be 0.15 ± 0.13 Mm^{-1}, which is consistent with our value. Absorption is low during winter as a result of factors such as elevation, sources, transport, and scavenging (Jaffrezo *et al.*, 1994). Notable peaks in absorption are observed in May in 2011, May and August in 2012, April and June in 2013, and April and August in 2014. These peaks may be related to forest fire burning plumes from northern parts of North America which have been observed to reach Summit as well as other parts of the Arctic (Jaffrezo *et al.*, 1998; Stohl *et al.*, 2006). The average measured σ_{ap} in spring is 0.19 ± 0.15 Mm^{-1} and 0.12 ± 0.14 Mm^{-1} in summer. The data suggests that

periods of high absorption occur in both spring and summer, with higher values most typical in the spring.

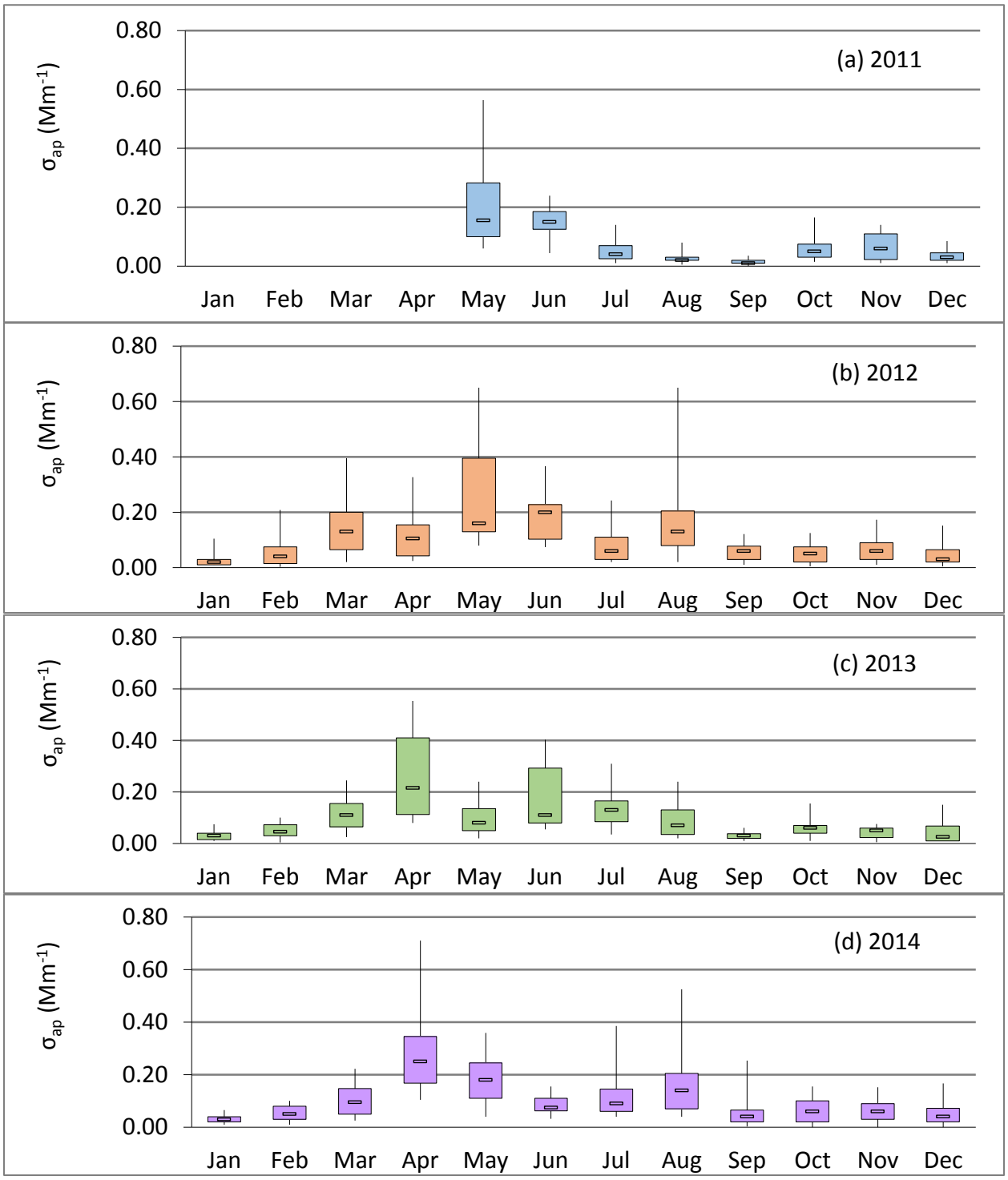

Figure 1. Monthly σ_{ap} at 550 nm at Summit for (a) 2011, (b) 2012, (c) 2013, and (d) 2014.

A study performed at Summit in 1993 showed that atmospheric concentrations of soluble ionic species (SO_4^{2-}, NO_3^-, Na^+, NH_4^+, Ca^{2+}, K^+, and Mg^{2+}) exhibited a similar seasonality (Bergin *et al.*, 1995). Concentrations of cation and anion aerosol species were both high in spring and summer, but spring exhibited higher values. Another study conducted on the GIS measured sulfate and methanesulfonic acid, resulting in the same seasonality (Jaffrezo *et al.* 1994). It suggested that the seasonality was caused by a combination of factors including transport and changes in meteorology which may explain our findings at Summit.

3.1.2 Scattering

Fig. 2 shows monthly scattering coefficient observed at Summit. Similar to absorption, scattering is typically highest during the months of April-August, with an average σ_{sp} of 2.35 ± 2.80 Mm^{-1}. Average σ_{sp} in the spring is slightly higher than the summer average, with values of 2.38 ± 2.03 Mm^{-1} and 2.32 ± 3.52 Mm^{-1}, respectively. The scattering coefficient co-varies with the absorption coefficient seasonally, exhibiting simultaneous periods of high values, with the exception of September, October, and November in 2014 when scattering was particularly high. Fig. 3 shows the correlation between σ_{sp} and σ_{ap}, displaying an R^2 value of 0.63.

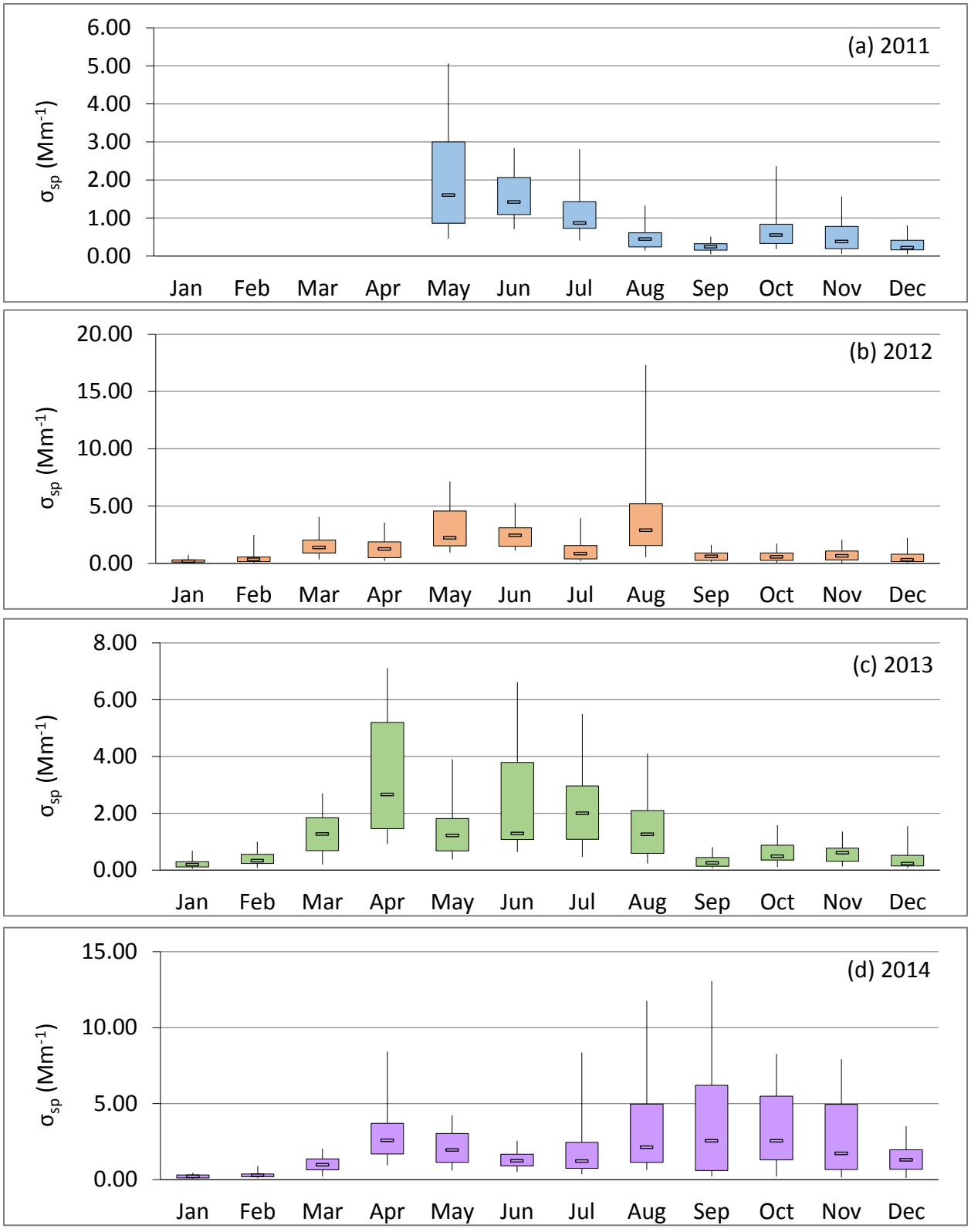

Figure 2. Monthly σ_{sp} at 550 nm at Summit for (a) 2011, (b) 2012, (c) 2013, and (d) 2014

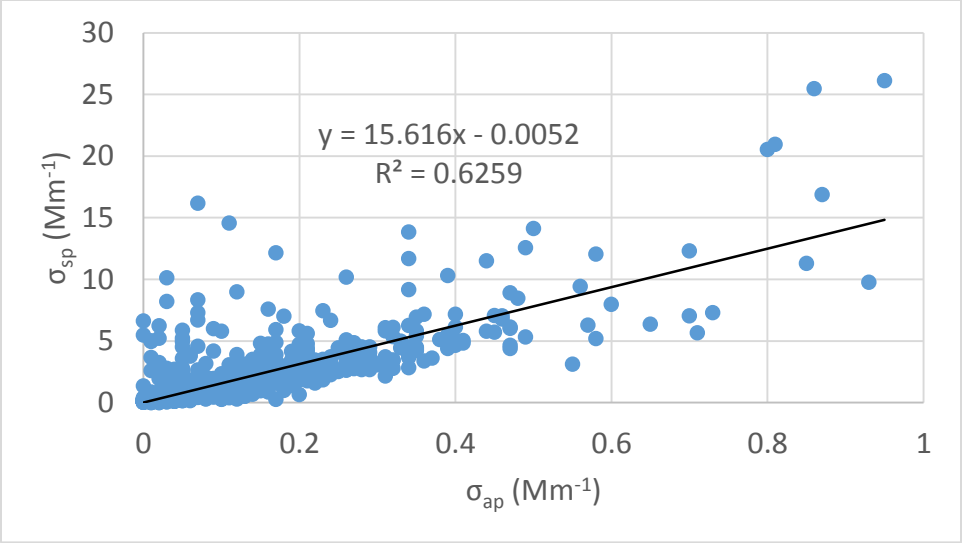

Figure 3. Relationship between σ_{ap} and σ_{sp} at 550 nm at Summit for the sampling period

3.1.3 Absorption and Scattering at Other Arctic Sites

The seasonal variation of σ_{ap} and σ_{sp} at Summit is different from that at other arctic sites. At Barrow, σ_{ap} and σ_{sp} are highest in the winter, likely due to high concentrations of submicron sulfate and sea-salt, and low during the summer (Delene and Ogren, 2002). Quinn *et al.* (2002) also show high σ_{ap} and σ_{sp} values in winter and low values in summer at Barrow, hypothesizing that small particles inefficient at scattering light, such as biogenic sulfur, are present during the summer. Another study shows that the seasonality of BC at Barrow, Ny-Ålesund, and Alert, Nunavut, Canada is characterized by high concentrations from Arctic haze during winter and spring while concentrations in the summer are low (Sharma *et al.*, 2013).

3.2 Single Scattering Albedo

Fig. 4 shows monthly values of single scattering albedo (at 550 nm) at Summit. Values during the winter and autumn months exhibit high variability due to both absorption and scattering coefficients being close to the detection limits of the nephelometer and CLAP. Single scattering albedo is observed to increase from spring to summer. This increase in ω_o may be due to an increase in light-scattering aerosols in the atmosphere such as methanesulfonic acid (MSA) during the summer when there is more sunlight (Bergin *et al.*, 1995; Jaffrezo *et al.*, 1994). Average spring and summer ω_o also show little interannual variability, with values between 0.93-0.94 and a 2011-2014 average of 0.93 ± 0.03. This value is compared with spring and summer ω_o at other Arctic sites such as Barrow and Ny-Ålesund and is summarized in Table 1.

Table 1. Summary of spring and summer aerosol optical properties over Summit, Barrow, and Ny-Ålesund. Summit values (at 550 nm) are averaged over May 8 to August 31 (2011) and April 1 to August 31 (2012-2014). Barrow values (at 550 nm) are averaged over April 1 to August 31 (2009-2013). Ny-Ålesund values (at 525 nm) are averaged over April 1 to August 31 (2005-2009). Values shown are: mean ± standard deviation (interquartile range)

Station	σ_{ap} (Mm^{-1})	σ_{sp} (Mm^{-1})	ω_o
Summit	0.15 ± 0.15 (0.14)	2.35 ± 2.80 (1.97)	0.93 ± 0.03 (0.03)
Barrow	0.16 ± 0.17 (0.19)	3.54 ± 4.10 (3.55)	0.94 ± 0.05 (0.05)
Ny-Alesund	0.27 ± 0.68 (0.18)	4.86 ± 5.36 (4.50)	0.95 ± 0.06 (0.03)

Single scattering albedo was calculated at Barrow to be 0.94 ± 0.05 and at Ny-Ålesund to be 0.95 ± 0.06. The value for ω_o at Summit is as low as those at Barrow and Ny-Ålesund despite the fact that it is an isolated high-altitude site, indicating that light absorption by aerosols are important even in the most remote Arctic locations.

Furthermore, Summit is snow-covered and reflective all throughout the spring and summer, while the other two sites are not. This suggests that aerosol light-absorption could be more significant in the more reflective region.

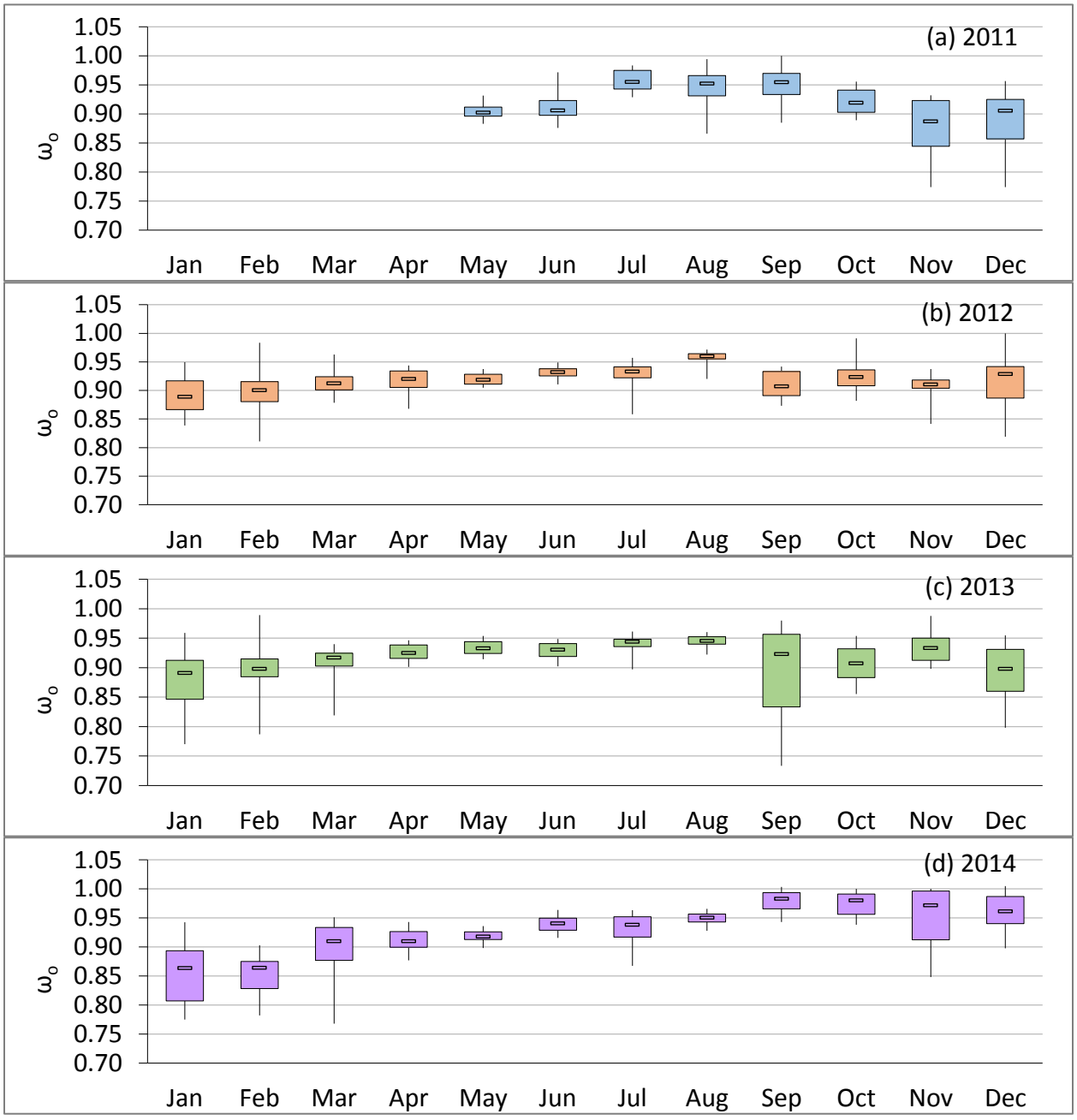

Figure 4. Monthly ω_o at 550 nm at Summit for (a) 2011, (b) 2012, (c) 2013, and (d) 2014.

3.3 Absorption Ångström Exponent

Figure 5 shows the monthly AAE observed at Summit. Spring and summer values show little interannual variability, averaging a value of 0.97 ± 0.29. This value suggests that BC, which has an AAE of 1.00 (Yang *et al.*, 2009), and not dust and/or BrC is the main light absorber throughout the sunlit seasons. Fig. 3 shows the frequency distribution of AAE measured during spring and summer. There is a peak in frequency at 0.9-1.0. This value typically indicates that the BC source is fossil fuel combustion, however, BC over Greenland has been suggested to originate from both fossil fuel and biomass burning (Hegg *et al.*, 2010). Studies show that biomass burning is an important source of BC at Summit (Jaffrezo *et al.*, 1998; Stohl *et al.*, 2006). Biomass burning produces both black carbon and brown carbon (Lee *et al.*, 2014) and has an AAE of 1.53 ± 0.10 (Yang *et al.*, 2009). This value is greater than 1.00 for just BC alone due to contributions from BrC. Fig. 5 exhibits a frequency of approximately 5% for AAE of 1.5-1.6, which is a weak indication of biomass burning. The BrC contribution to AAE is not apparent in the data due possibly to aging in the atmosphere. Studies show that BrC can become less light absorbing via sunlight bleaching (Zhong and Jang, 2014) and also via photolysis (Lee *et al.*, 2014). Thus, it is inconclusive what the sources of BC over this region are.

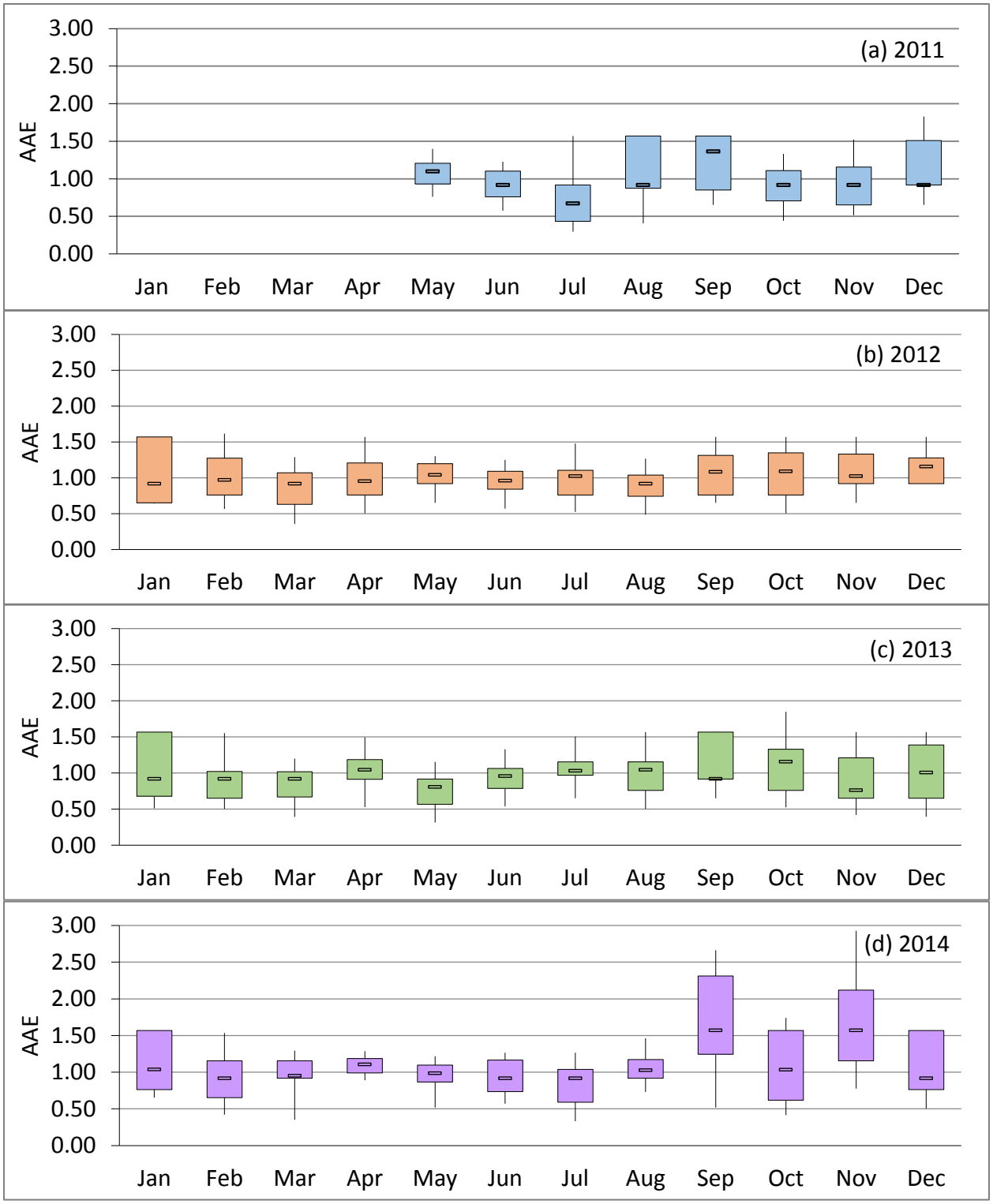

Figure 5. Monthly AAE (450-700 nm) at Summit for (a) 2011, (b) 2012, (c) 2013, and (d) 2014.

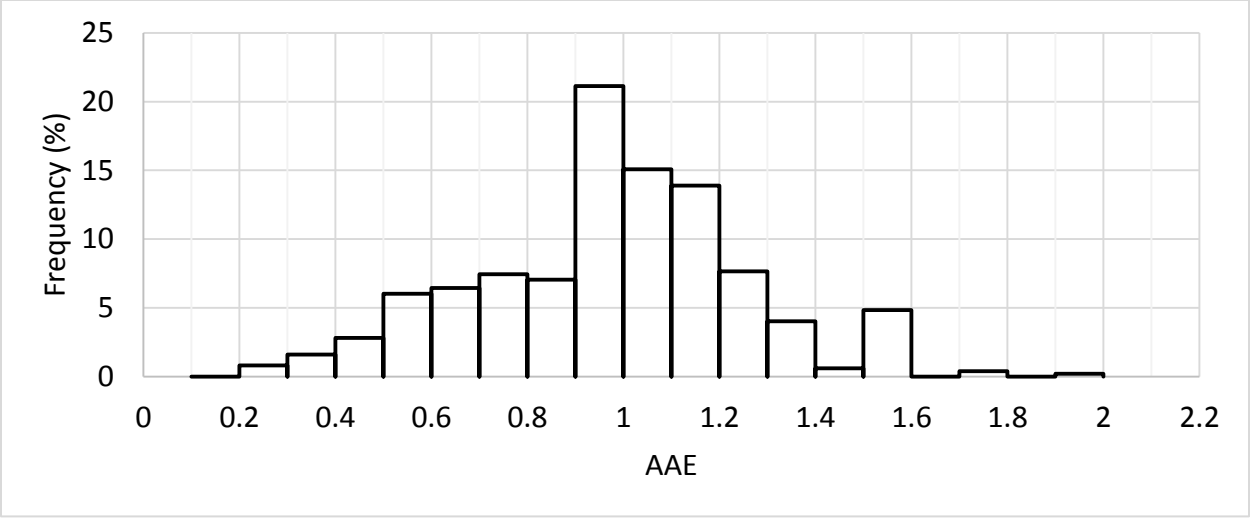

Figure 6. Frequency distribution of absorption Ångström exponent (450-700 nm) at Summit during spring and summer, 2011 to 2014

CHAPTER 4

CONCLUSIONS

This study, for the first time in literature, presents continuous, interannual measurements of the aerosol optical properties (σ_{ap}, σ_{sp}, AAE, ω_o) needed to characterize aerosol seasonality and interannual variability as well as identify the main light-absorbing aerosol over central Greenland. Several major findings are made from this study. Absorption and scattering are typically low during winter and autumn and highest during the sunlit spring and summer months. While peaks in absorption are observed in both spring and summer, values in the spring are typically higher. Scattering maximums seem to vary between spring and summer with each year with the exception of 2014 when the highest values were in autumn. AAE during these periods have a mean value of 0.97 ± 0.29, which indicate that the main source of light absorption is BC and not dust or BrC. While we can identify BC as the main light absorber, it is uncertain what source it originated from, but evidence suggests it could be from biomass burning. Single scattering albedo is higher in the summer than in the spring. Mean ω_o during spring and summer has a value of 0.93 ± 0.03, as low as other less-reflective Arctic sites. Overall, the data help improve our understanding of the seasonality and sources of aerosols over central Greenland. Furthermore, the results indicate that aerosol light absorption is significant even in the most remote parts of the Arctic.

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